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PATENT SPECIFICATION

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(54) NAPHTHOLACTAM DYES

(71) We, BASF AKTTENGESELL-SCHAFT, a German Joint Stock Company, of 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The invention relates to dyes of the formula 10 (I):

in which

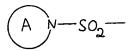
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R¹ is hydrogen, chloro, bromo, alkyl, alkoxy, nitro or arylmercapto;

5 R2 is hydrogen or chloro;

Ra is hydrogen, chloro, bromo, alkyl, alkoxy, nitro, carboxylic acylamino, alkylsulfonylamino, arylsulfonylamino, alkylsulfonyl, arylmercapto, arylsulfonyl, alkylsulfonyl, unsubstituted or substituted sulfamoyl, alkanoyl, aroyl or a heterocyclic group of the formula:—



in which the ring A is a saturated heterocyclic group;

R⁴ is hydrogen, chloro, alkoxy or arylmercapto; or

R³ and R⁴ together form a radical of the formula:

R⁵ is hydrogen, chloro or alkoxy; and R⁶ and R⁷ independently of one another are

alkyl; alkyl bearing hydroxy, cyano, alkoxy or carbalkoxy as a substituent; phenyl; or phenyl bearing cholro, methyl or methoxy as a substituent, at least one of R¹ to R⁵ being different from hydrogen.

Interpretations of R¹ to R⁵:

It is to be understood that references herein to aryl moieties (e.g. in arylmercapto or in aroyl) are intended to cover substituted aryl moieties as well as unsubstituted aryl moieties.

Examples of alkyl, alkoxy and arylmercapto groups for R¹ are methyl, ethyl, methoxy, ethoxy, phenylmercapto or phenylmercapto bearing chloro, methyl, methoxy, phenyl, phenoxy or methoxycarbonyl as a substituent.

Examples for R⁸ are the same radicals as for R1 and also naphthylmercapto, acetylaminophenylmercapto, acetylamino, propionylamino, benzoylamino, benzoylamino bearing chloro, methyl or methoxy as a substituent, ethylsulfonylamino, methylsulfonylamino, tolylsulfonylamino, phenylsulfonylamino, methylmercapto, β - hydroxyethylmercapto, methylsulfonyl, ethylsulfonyl, phenylsulfonyl, tolylsulfonyl, chlorophenylsulfonyl, acetyl, propionyl, butyryl, benzoyl, benzoyl bearing methyl, methoxy, chloro or bromo as a substituent, sulfamoyl, N - methylsulfamoyl, N ethylsulfamoyl, N - butylsulfamoyl, N - phenylsulfamoyl, N - chlorophenylsulfamoyl, N - methylphenylsulfamoyl, N - methoxyphenylsulfamoyl, N - trifluoromethylphenylsulfamoyl, N - methyl - N - phenylsulfamoyl, N,N - dimethylsulfamoyl, N,N - diethylsulfamoyl, N,N - dipropylsulfamoyl, N,N - dibutylsulfamoyl, pyrrolidinosulfonyl, piperidinosulfonyl or morpholinosulfonyl. Radicals of the formulae: NHCONH2, NHCONHCH3 and NHCONHC.H. are also suitable,

Examples of radicals R⁴ are hydrogen, chloro and the alkoxy and arylmercapto radicals specified for R¹.

Examples of alkoxy radicals R⁵ are methoxy and ethoxy.

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Particular examples of R⁶ and R⁷ are alkyl of one to four carbon atoms, alkoxyalkyl of three to eight carbon atoms, hydroxyalkyl of two or three carbon atoms, cyanoethyl, alkoxy-

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carbonylalkyl of one to four carbon atoms in the alkoxy and also phenyl.

Specific examples are: propyl, butyl, methoxyethyl, ethoxyethyl, butoxyethyl, methoxypropyl, ethoxypropyl, pentoxypropyl, β -hydroxyethyl, β -hydroxypropyl, methoxycarbonylethyl, ethoxycarbonylethyl and butoxycarbonylethyl and preferably methyl, ethyl and phenyl.

Dyes of the formula (I) may be prepared by reacting a naphtholactam of the formula (II):

or a derivative of the same in which the 15 grouping

is replaced by

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with an N,N'-disubstituted barbituric acid 25 derivative of the formula (III)

Compounds of the formula (II) are known from the literature or may be prepared by methods analogous to those described in the 30 literature.

The reaction of the naphtholactams with the compounds of formula (III) proceeds in the presence of a condensing agent; when the said naphtholactam derivatives are used the presence of a condensing agent may be dispensed with.

Phosphorus halides such as phosphorus

pentachloride, phosphorus trichloride or phosphorus oxytribromide and particularly phosphorus oxytrichloride are suitable as condensing agents.

The reaction may be carried out in an inert solvent such as toluene, chlorobenzene, dichlorobenzene, nitrobenzene or dioxane or also in an excess of the condensing agent.

When the reaction is carried out with a naphtholactam derivative and without a condensing agent the abovementioned solvents are again suitable; examples of additional solvents are pyridine, glacial acetic acid, dimethylformamide and N-methylpyrrolidone.

The reactions are known in principle and details may be taken from the Examples in which parts and percentages are by weight.

The invention relates particularly to dyes of the formula:

R1 is hydrogen, chloro, bromo, C1 to C4 alkyl, methoxy, ethoxy, nitro, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

R2 is hydrogen or chloro;

R3 is chloro, bromo, C1 to C4 alkyl, methoxy, ethoxy, nitro, acetylamino, propionylamino, - benzoylamino, methylsulfonylamino, ethylsulphonylamino, phenylsulfonylamino, tolylsulfonylamino, methylmercapto, ethylmerphenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, naphthylmercapto, phenylsulfonyl, phenylsulfonyl bearing chloro or methyl as a substituent, methylsulfonyl, ethylsulfonyl, N - mono - C_1 - to - C_4 - alkyl - substituted sulfamoyl, N_1N - di - C_1 - to - C_8 - alkyl substituted sulfamoyl, N - phenylsulfamoyl, N - chlorophenylsulfamoyl, N - methylphenylsulfamoyl, N - methoxyphenylsulfamoyl, N - trifluoromethylphenylsulfamoyl, pyrrolidonosulfonyl, piperidinosulfonyl, morpholinosulfonyl, C₂ to C₄ alkanoyl, benzoyl, or benzoyl bearing chloro, bromo, methyl, ethyl, methoxy or ethoxy as a substituent;

is hydrogen, chloro, methoxy, ethoxy, phenylmercapto, or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

R⁸ and R⁴ together are

R5 is hydrogen, chloro, methoxy or ethoxy;

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R° and R′ are independently C₁ to C₄ alkyl, C₂ to C₈ alkyl bearing hydroxy, cyano, C₁ to C₄ alkoxy or C₁ to C₄ alkoxycarbonyl as a substituent, phenyl, or phenyl bearing chloro, methyl or methoxy as a substituent. Particular industrial importance attaches to dyes of formula (Ia):

$$B^{1} \xrightarrow{\text{RN}-C} N^{-B^{3}} \text{NZ}_{B}^{1} \text{O}$$
 (Ia)

in which 10 B1 is hydrogen, chloro, bromo or arylmer-

B2 is chloro, bromo, arylmercapto, arylsul-

fonyl, or aroyl; and B3 and B4 are independently methyl, ethyl or phenyl, methyl being particularly preferred.

The following are preferred arylmercapto, arylsulfonyl and aroyl radicals: phenylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, phenylsulfonyl or benzoyl, or benzoyl or phenylsulfonyl bearing chloro or methyl as a substituent.

Dyes of formula (I) are yellow to violet and have high brilliance and color strength. They are suitable for dyeing synthetic fibers, particularly polyester fibers, and dyeings having very good fastness properties and particularly excellent fastness to light are obtained. Thermal resistance properties may be influenced by the choice of substituents. Dyes which sublime easily are very suitable for transfer printing.

The new dyes are moreover eminently suitable for the mass coloration of plastics; fluorescent colorations having high fastness to light are obtained as a rule.

The invention includes within its scope dye formulations for dyeing synthetic fibers, the formulations containing a dye in accordance with the invention.

40 The following Examples illustrate the inven-

Example 1.

200 parts of phosphorus oxychloride is dripped into a mixture of 309 parts of 4-phenylsulfonylnaphtholactam - (1,8), 165 parts of N,N'-dimethylbarbituric acid and 1200 parts by volume of toluene at 90°C. The whole is stirred for 6 hours at 100°C and 1200 parts by volume of methanol is added during cooling so that the dye is deposited in the form of brown crystals. After cooling, the dye is filtered off, washed with methanol and dried. 357 parts of the dye of the constitution

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is obtained which dyes polyester from an aqueous liquor brilliant yellow hues of high tinctorial strength and very good light fastness and thermal stability.

Example 2.

The procedure of Example 1 is followed, but the N,N'-dimethylbarbituric acid is replaced by an equivalent amount of N - methyl - N - phenylbarbituric acid. 427 parts of the dye of the constitution

is obtained which dyes polyester fast golden yellow hues.

Example 3.

273 parts of 4 - benzoylnaphtholactam - 70 (1,8) and 190 parts of N,N'-dimethylbar-bituric acid are introduced into 1000 parts by volume of toluene and stirred at 95°C. 300 parts of phosphorus oxychloride is dripped in within thirty minutes so that the starting 75 materials pass into solution. The whole is stirred for four hours at 100°C and during cooling 2000 parts by volume of methanol is allowed to flow in. After suction filtration, washing with methanol and drying there is obtained 360 parts of the dye having the constitution:

in the form of yellowish brown crystals. The dye dyes polyester brilliant greenish yellow 85 shades from an aqueous liquor; the dyeings have very good fastness to dry-heat pleating and setting and to light. In thermoplastics such as polystyrene fluorescent yellow hues having outstanding stability at high temperatures and outstanding fastness to light are obtained.

Example 4.

250 parts of phosphorus oxychloride is added at 100°C within one hour to a mixture of 277 parts of 4 - phenylmercaptonaphtholactam - (1,8), 190 parts of N,N'-dimethylbarbituric acid and 900 parts by volume of chlorobenzene. The whole is stirred for another four hours at 100°C and during cooling there is added a solution of 150 parts of triethylamine in 1500 parts by volume of ethanol. After suction filtration, washing with ethanol and drying there is obtained 349 parts of the dye of the constitution:

in the form of a dark red crystalline powder. Brilliant scarlet dyeings and prints of a high level of fastness properties are obtained on polyesters. Thermoplastics such as polystyrene for example are colored fluorescent scarlet shades having excellent stability at high temtemperatures and excellent fastness to light.

Example 5.

293 parts of 4 - phenylmercaptothionaphtholactam - (1,8) and 160 parts of N,N' dimethylbarbituric acid are boiled in 400 parts
by volume of N-methylpyrrolidone for eight
hours. 200 parts by volume of ethanol is added
during cooling. The product is suction filtered,
washed and dried. 302 parts of dye is obtained which is identical with that specified
in Example 4.

Example 6.

307 parts of 4 - phenylmercapto - thiomaphtholactam - (1,8) - S - methyl ether and 160 parts of N,N'-dimethylbarbituric acid are boiled in 600 parts by volume of acetic acid for 1 hour. The dye precipitates in the form 25 of red crystals. Upon working up, 328 parts of dye is obtained which is identical with that specified in Example 4.

Example 7.

41.3 parts of 2,4 - bis - (4 - methylphenyl30 mercapto) - naphtholactam - (1,8) and 20
parts of N,N'-diethylbarbituric acid are introduced into 150 parts by volume of dichlorobenzene and stirred at 95°C. 30 parts of
phosphorus oxychloride is dripped in so that
35 the components pass into solution. The whole
is stirred for six hours at 95°C, diluted with
300 parts by volume of methanol and allowed
to cool while stirring. The red crystals are

suction filtered, washed with methanol and dried. 49.2 parts of the dye of the constitu- 40 tion:

is obtained which gives in polystyrene bluish red hues of outstanding stability at high temperatures and outstanding fastness to light.

Example 8. 268 parts of the compound of the formula

(prepared by the method of German Laid-Open Specification 1,445,624, Example 2) is heated for 30 minutes at 100 to 120°C in 1,000 parts of xylene. 220 parts of N - methyl - N' - (3 - methoxypropyl) - barbituric acid is added and heating is continued at 100°C for 120 minutes. During cooling, 500 parts of methanol is added and the dye isolated in the usual manner. 311 parts of the dye of the constitution

is obtained in the form of orange crystals, which dye polyester reddish yellow hues having very good lightfastness.

The following dyes of the general formula (I) are prepared according to the methods specified in Examples 1 to 8.

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Hue	greenish yellow	:	reddish yellow	:	:	:	2			orange yellow	:	yellowish orange	:		:	reddish yellow			:	
	gree	•	redd	•	-	•	•		-	огаг		yell				redo				
R'	-CH3	:	2	-C,H,	:	-CH3	-C ₂ H _s	-(CH ₂)30CH ₃	-CH,	a	-C,H,	-CH3	-C,H,	-(СН ₂),ОС,Н	-CH,	-CH3	-C,H,	-(CH ₂) ₃ OCH ₃	-C,H,	-CH,
R°	-CH3	•	:	:	-C,H,	-CH3	:	Ę	-CH	:	-C ₂ H ₅	-CH	-C,H,	Ę.	-C ₂ H ₅	-CH	:	=	-C,H,	-CH,
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R	-C ₂ H _s	Ħ	•	:	:	ם	:	ວ	Ħ	\$		î.	:	:	:	:	2	:		Ŗ
Ex.	6	10	11	12	13	14	15	16	.17	18	19	20	21	22	23	24	25	26	27	28

	Hue	reddish yellow	vellowish orange					golden yellow		ih ye	golden yellow	yellow	•	**	•	:	: :	:		
	R,	-CH3		—(СН ₂),ОСН,	-(CH ₂),0C ₂ H ₃	-C,H,	-C,H,	-CH3	-C _. H _s	-CH ₃		:	l,	-C,H,	-CH,	2	-C,H,	2	2	-СН,
	ఙ	-CH,	•	:		-C,H,	-C,H	-CH	-C ₂ H ₅	-CH	:	2	-CH	-C,H,	-CH3	2	-C,Hs	:		-CH3
nued)	Rs	H	:	:	:	:	:	:	2			:	:	:	:	2	:	:	:	:
TABLE (Continued)	R4	Н	:	:	:	:	- 2	:	:	:	:	2	:	:		•	2	:	:	2
T/	R,	Br	2	*		:	:	NO,	:	Br	NO ₂	осн	<u>,</u>		0C2Hs	NHCONH ₂	*	NHCONIICH3	NHCONHC,H,	NHCOC,H,
	R²	H	:	:	\$	2	:	H	:	:	\$	н	2	:	:	2	5	•	:	:
	R.	ਹ	Br.	•	:	:	\$	н	2	NO ₂	Br	H	ç	:	•	:	2	\$	•	:
	Ex.	29	30	31	32	33	34	35	36	37	38	39	40	41	42	43	44	45	46	47

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	Hue	yellow	•	•	•	•	:	orange	yellowish red	:		:	*.	orange yellow	:	-		orange		
	H	yel	•	•	. •	•	•	ora	yej					ога		red	2	ora	red	
	R7	$-C_2H_s$	-CH,	2		•	:	:	$-C_2H_s$	-(CH ₂),0CH ₃	-C _j H _s	C,H,	-C,H,	-CH3	-(CH ₂) ₃ OCH ₃	-CH3	-C ₂ H _s	-CH,	=	-C,H,
	R ⁶	-C ₂ H _s	-CH	:	:	2	:	:	2	•	-C ₂ H ₅	-CH,	-C,H,	-CH3	2	-CH3	-C ₂ H ₅	-CH ₃	•	:
ed)	R⁵	Н	2	:	=	-0CH3	-0C ₂ H ₅	н	:	:	=	2	•	:	2	•	=		2	
ABLE (Continued)	R4	Н	:	:	-0CH3	H	2	:	:	:	•	•	•	SC,H	:	×	:	SC,H,CH, (*)	Ħ	•
	R³	NHCOC,H	NHSO ₂ C ₆ H ₅	NHSO,CH,	H	.	2	SCH,	SC,H,	SC,H,	:	:	•	· C		SC,H,CH, (*)	:	C	SC,H,OCH, (*)	SC,H,OCH,.(*)
	R²	H	:	:	*	:	:	:	:	2	2	:	*	:	:	2	=	:	•	
	ጁ	Н	:	:	:	:	2	2	:	2	:	2	:	•	. :	:	:	:		
	Ex.	48	49	20	51	22	53	54	55	99	23	58	89	09	61	62	63	64	99	99

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Hue	red	yellowish red	orange	:	yellowish orang	red	. =	yellowish red	red	м		orange	:		reddish orange				yellowish red
R,	-(CH ₂) ₃ OC ₂ H ₆	-CH,	:	-C,H,	СН,		:	:		2	-C ₂ H ₅	ĠĦ,	-C ₄ H,	-CH ₃	=	-C ₄ H,	-(CH ₂)30C ₂ H ₅	-C,H,	-CH,
ጼ	-CH3	. A	2	:	\$	2	:	*	2		-C ₂ H ₅	-CH,	2		:	-C,H,	Ę,	-C,H,	-CH3
Rs	н	2	:	:	:	:	:	=	:	•	:	2	â	:	:	2	=	:	:
R4	Н	SC,H,OCH, (*)	æ	•	•	:	2	:	•	:	:	2	=	•		:	â	2	SC,H,
R³	SC,H4OCH3 (*)	<u></u>	SC,H,CI (*)	:	SC,H,Cl, (2,5)	SC,H,C,H, (*)	SC,H,OC,H, (*)	SC,H,NHCOCH, (*)	SC,H,C00CH, (*)	Naphthyl-2-S-	e.	Ü	2	NO ₂ .	SC,H5	:	:		
R2	H	:	:	. =	:	:	2	:	:	•	:	:	:	:	•	2	2	:	:
R	Н	:	£		2	:	2	2	2	•		$SC_{\nu}H_{\mu}$:	*	•		•	•	H
Ex.	<i>L</i> 9	89	69	. 0/	11	72	73	74	75	92.	11	78	19	80	81	82	83	84	8.5
	R ¹ R ² R ³ R ⁴ R ⁵ R ⁶ R ⁷	R¹ R² R³ R° R³ R° H H SC ₀ H ₂ OCH ₃ (*) H H -(CH ₂) ₂ OC ₂ H ₅ red	R¹ R³ CCH3, CC2, H³ CCCH3, CC2, H³ CCCH3, CC2, H³ CCCC2, CC2, H² CCCC2, H² CCCC2, H² CCC2, H² CC	R¹ R³ CCH₃, DCC₂H₅ R³ CCH₃, DCC₂Hӻ R³ CCH₃ CCCH₃ CCCH₃ CCCCH₃ CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC	R¹ R² R³ CCH₂,3OC₂H₅ R³ CCH₂,3OC₂H₅ R³ CCH₃ CCCH₃ CCCCH₃ CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC	R¹ R³ CCH₃, CC2,H₅ CCH₃, CC2,H₅ R³ CCH₃, CC3,H₂ CCH₃, CC3,H₂ R³ CCH₃, CC3,H₂ R³ CCH₃, CC3,H₂ R³ CCH₃, CC4,H₂ R³ CC4,H₂	R¹ R³ CCH₂₃,0C₂H₅ R³ CCH₃, CCCH₃, CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC	R¹ R³ CCH₂₃,0C₂H₅ R³ CCH₃, CCCH₃, CCCH₃, CCCH₃, CCCH₃, CCCCCCCT, CCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCCC	R¹ R³ CCH₃, 3,0C₂H₂, 8 R³ CCH₃, 2 CCH₃, 2 R³ R³ R³ </td <td>R¹ R³ CCH₂₃,0C₂H₂, R³ CCH₃, CCH₃</td> <td>R¹ R³ CCH₂₃³0C₂H₅ R³ CCH₂₃³0C₂H₅ R³ CCH₃³ CCH₃³ CCH₃³ R³ CCH₃³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ R° CCH₃³ R³ R° R°</td> <td>R¹ R³ R³<</td> <td>R¹ R³ R³<</td> <td>R1 R2 R3 R4 R5 R6 R7 H SC6H4OCH3 (**) H —CH3,30C2H6 11 CI SC6H4OCH3 (**) 10 —CH3,30C2H6 12 SC6H4CI (**) H 10 —CH3,30C2H6 12 —CH4,30C2H6 —CH4,30C2H6 13 SC6H4CI (**) —CH4,30C2H6 14 SC6H4CI (**) 15 SC6H4CI (**) </td> <td>R¹ R³ R³<</td> <td>R¹ R³ R³<</td> <td>R¹ R³ R³<</td> <td>R¹ R³ R³<</td> <td>H R³ R³<!--</td--></td>	R¹ R³ CCH₂₃,0C₂H₂, R³ CCH₃, CCH₃	R¹ R³ CCH₂₃³0C₂H₅ R³ CCH₂₃³0C₂H₅ R³ CCH₃³ CCH₃³ CCH₃³ R³ CCH₃³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ CCH₃³ R³ R° CCH₃³ R³ R° R°	R¹ R³ R³<	R¹ R³ R³<	R1 R2 R3 R4 R5 R6 R7 H SC6H4OCH3 (**) H —CH3,30C2H6 11 CI SC6H4OCH3 (**) 10 —CH3,30C2H6 12 SC6H4CI (**) H 10 —CH3,30C2H6 12 —CH4,30C2H6 —CH4,30C2H6 13 SC6H4CI (**) —CH4,30C2H6 14 SC6H4CI (**) 15 SC6H4CI (**)	R¹ R³ R³<	R¹ R³ R³<	R¹ R³ R³<	R¹ R³ R³<	H R³ R³ </td

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Ex.	R¹	R ²	R³	R	Rs	ጼ	R,	Hue
98	Н	н 🦠	SC,H,	SC,H,	H	-CH ₃	-C,H,	yellowish red
87	SC, H,CH, (*)	•	SC,H,CH, (*)	H	:	:	-CH,	red
88		٠ :	à	Н	:	-CH ₃	-(CH ₂) 10CH ₃	2
68		•	c	н	2	_C,H,	-C,H,	
90	SC,H,OCH, (*)	:	SC,H,OCH, (*)			. —CH,	-CH,	bluish red
91	2	2	:	:	:	-C ₂ H ₅	-C ₂ H _s	
92	н	:	:	SC,H,OCH, (*)		-CH³	-сн,	yellowish red
93	SC,H,CI (*)	2	SC ₈ H ₄ Cl (4)	н	2	:	a	reddish orange
94	SC,H,CI (*)	2	•		2	:	-C,H,	2
95	SC,H,CI, (2,5)	•	SC,H,C1, (2,5)	66	:	:	-CH ₃	orange

Example 96.
200 parts of N,N'-dimethylbarbituric acid and 275 parts of the compound of the constitution:

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are brought into solution in 1800 parts by 1 volume of anhydrous nitrobenzene by heating. 220 parts of phosphorus oxychlouide is dripped in at 100°C and the whole is stirred for another twelve hours at 100°C. After cooling it is diluted with an equal volume of ethanol, suction filtered, washed with ethanol and edried. 321 parts of the dye of the formula: 9

is obtained in the form of a dark crystalline powder. When the dye is incorporated into polystyrene it gives violet hues having good stability to high temperature and good lightfastness properties.

When the N.N-dimethylbarbituric acid is replaced by barbituric acids having other substituted by barbituric acids having other substitutents, dyes having very similar hues are obtained analogously to Examples 1 to 8, are:—

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Example	$R^{3}(R^{1}=R^{2}=R^{4}=R^{5}=H)$	R ⁶	R ⁷	Hue
97	–SO₂C ₆ H ₅	-CH ₃	–C₄H,	golden yellow
98	11	**	-(CH ₂) ₃ OC ₂ H ₅	33 33
99	**	-C ₄ H ₉	-C ₄ H ₉	22
100	$-SO_2C_6H_4CH_3$ (4)	-CH ₃	-CH ₃	,, ,,
101	"	,,	$-C_2H_5$	** **
102	,,	$-C_6H_5$	-C ₆ H ₅	,, ,,
103	–SO₂C₅H₄Cl	-CH ₃	-CH ₃	,,
104	**	$-C_2H_5$	$-C_2H_5$,, ,
105	**	CH ₃	-C₄H,	"
106	**	-C ₄ H ₉	-C,H,	33 33 .
107	$-SO_2N(CH_3)_2$	-CH ₃	-CH ₃	yellow
108	$-SO_2N(C_2H_5)_2$,,	,,	**
109	$-SO_2N(C_4H_9)_2$,,	,	**
	\sim			
110 111	-so ₂ zi			•
112	-so ₂ N	-	-с ₂ н ₅ -сн ₃	
113	-so _z n	-c4H9	-c4H9	
114	-SO ₂ NHC ₆ H ₅	-CH ₃	-CH ₃	
115	-SO ₂ NHC ₆ H ₄ CH ₃ (4)	,,	,,	**
116	$-SO_2NHC_6H_4C1$ (2)	,,	"	> 2
117	-SO₂NHC₀H₄OCH₃ (4)	,,	39	37
118	$-SO_2NHC_6H_4CF_3$ (3)	,,	"	19
	CH₃			
119	–SO₂N	,	>>	**
	C ₆ H ₅			
120	-COCH ₃	,,		greenish yellow
121	-COC ₃ H ₇ (iso)	-C ₆ H ₅	-C ₆ H ₅	33 33
122	-COC ₆ H ₅	-C ₂ H ₅	-C _z H ₅	» »
123	**	-CH ₃	-C₄H,	2) 29
124	**	,,	-C ₆ H ₅	** **
125	29	−C₅H₅	,,	37 37

Exampl	e $R^{3}(R^{1}=R^{2}=R^{4}=R^{5}=H)$	R ⁶	R ⁷	Hue
126	-COC ₆ H ₄ CH ₃ (2)	-CH ₃	-CH ₃	yellow
127	-COC ₆ H ₄ CH ₃ (4)	,,	**	**
128)	, ,,	-C ₂ H ₅	**
129	-COC ₆ H ₄ Cl (2)	,,	-CH ₃	greenish yellow
130	-COC ₆ H ₄ CI (4)	,,	**	,,, ,,,
131	29	$-C_2H_5$	$-C_2H_5$	13 27
132	"	-CH ₃	-(CH ₂) ₃ OCH ₃	33
133	-COC,H3Cl2 (2,4)	**	-CH ₃	23 23
134	,,,	. ,,	-C₄H ₉	» »
135	-COC ₆ H ₃ Cl ₂ (3,4)	17	-CH ₃	23
136	-COC ₆ H ₃ Cl ₂ 2,5)	,,		» » .
137	***	**	$-(CH_2)_3OC_2H_5$	33 . 33
138	,,	-C ₆ H ₅	−C ₆ H ₅	. ,, ,,

in which

Example 139.

208 parts of N - methyl - N' - butylbarbituric acid and 271 parts of the compound of the constitution:

are stirred into 1200 parts by volume of anhydrous trichlorobenzene at 100°C. 300 parts of phosphorus oxychloride is dripped in within one hour and the whole is stirred for another eight hours at 100°C to 110°C. During cooling the whole is diluted with 800 parts by volume of methanol and then suction filtered, washed with methanol and dried. 361 parts of the dye of the constitution:

is obtained in the form of reddish brown crystals. Luminous orange colorations having good fastness properties are obtained with the dye in thermoplastics, as for example polystyrene. WHAT WE CLAIM IS:—

1. A naphtholactam dye of the formula:

R¹ is hydrogen, chloro, bromo, alkyl, alkoxy, nitro or arylmercapto;
R² is hydrogen or chloro;
R³ is hydrogen, chloro, bromo, alkyl, alkoxy, nitro, carboxylic acylamino, alkylsulfonylamino, arylsulfonylamino, alkylmercapto,

amino, aryisulfonylamino, alkylmercapto, aryimercapto, aryisulfonyl, alkylsulfonyl, sulfamoyl, sulfamoyl bearing one or two substituents on the nitrogen atom, alkanoyl, aroyl or a heterocyclic group of the formula:—

$$\bigcirc \text{A} \text{N-so}_2 -$$

in which the ring A is a saturated heterocyclic group;
R⁴ is hydrogen, chloro, alkoxy or arylmercapto; or
R³ and R⁴ together form a radical of the formula:

R⁵ is hydrogen, chloro or alkoxy; and R⁶ and R⁷ are independently alkyl, alkyl bearing hydroxy, cyano, alkoxy or carbalkoxy as a substituent, phenyl or phenyl bearing chloro, methyl or methoxy as a substituent;

and at least one of the radicals R¹ to R⁵ is other than hydrogen.

10 2. A dye of the formula:

n which

25

30

35

R¹ is hydrogen, chloro, bromo, C₁ to C₄ alkyl, methoxy, ethoxy, nitro, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent;

R² is hydrogen or chloro;
R³ is chloro, bromo, C₁ to C₄ alkyl, methoxy, ethoxy, nitro, acetylamino, propionylamino, benzoylamino, methylsulfonylamino, ethylsulfonylamino, phenylsulfonylamino, tolylsulphonylamino, methylmercapto, ethylmercapto, phenylmercapto bearing chloro, methyl or methoxy as a substituent, naphthylmercapto, phenylsulfonyl, phenylsulfonyl bearing chloro or methyl as a substituent, methylsulfonyl, ethylsulfonyl, N - mono - (C₁ to C₈) - alkylsubstituted sulfamoyl, N,N - di - C₁ to C₈) - alkyl-substituted sulfamoyl, N - phenylsulfamoyl, N - methylphenylsulfamoyl, N - methylphenylsulfamoyl, N - methylphenylsulfamoyl, N - trifluoromethylphenylsulfamoyl, pyrrolidinosulfonyl, piperidinosulfonyl, morpholinosulfonyl, C₂ to C₄ alkanoyl, benzoyl, or benzoyl bearing chloro, bromo, methyl, ethyl, methoxy

R4 is hydrogen, chloro, methoxy, ethoxy, phenylmercapto or phenylmercapto bearing chloro, methyl or methoxy as a substituent; or

R⁸ and R⁴ together are:

or ethoxy as a substituent;

45 R⁶ is hydrogen, chloro, methoxy or ethoxy; and R⁶ and R⁷ are independently C₂ to C₈ alkyl, C₂ to C₈ alkyl bearing hydroxy, cyano, C₁ to C₄ alkoxy or C₁ to C₄ alkoxycarbonyl as a substituent, phenyl or phenyl bearing chloro, methyl or methoxy as a substituent.

3. A dye as claimed in claim 1 and having the formula:

in which

55

B¹ is hydrogen, chloro, bromo or arylmercapto;

B² is chloor, bromo, arylmercapto, arylsulfonyl or aroyl; and

B³ and B⁴ independently are methyl, ethyl 6 or phenyl.
4. A dye as claimed in claim 1 and speci-

 A dye as claimed in claim 1 and speci fied in any one of the foregoing Examples.

 A process for the production of a dye as claimed in claim 1 wherein a naphtholactam.
 65 of the formula:

or a derivative of the same in which the grouping

is replaced by

or

is reacted with an N,N-disubstituted barbituric acid derivative of the formula:

6. A process as claimed in claim 5 carried out substantially as described in any one of the foregoing Examples.

7. A dye as claimed in Claim 1 and obtained by a process as claimed in Claim 5 or Claim

8. A dye formulation for dyeing synthetic fibers and which contain a dye as claimed in any of claims 1 to 4 or in Claim 7.

9. Plastics material whenever coloured by the presence therein of a dye as claimed in any of claims 1 to 4 or in Claim 7.

10. Synthetic fibers which have been dyed with a dye formulation as claimed in claim 8.

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